Radioactive Background in Hydrosphere Prior to Planned Extension of Nuclear Power Plant

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Abstract

Artificial radionuclides occurred in samples of surface water and other components of the hydrosphere in the last century. Their main sources were atmospheric tests of nuclear weapons and in Europe, the accident at the Chernobyl nuclear reactor. These radionuclides include mainly tritium, strontium 90 and caesium 137 in present.

In the area of concern, a power plant with two 1000 MW blocks has been in operation since 2001. At present, its extension is discussed. Regarding the assessment on the impacts of nuclear facilities which represent point sources of artificial radionuclides, it is necessary to take into account their impact on the hydrosphere.

The paper describes the development of artificial radionuclides activity concentrations in the recipient of the nuclear power plant waste water and its tributaries, uninfluenced by the power plant. The activity concentrations of caesium 137 and strontium 90 are permanently decreasing during the monitored period 1990-2012. Activity concentrations of tritium, detected in river sites not affected by Temelín nuclear plant, are slowly decreasing and their values are substantially lower than those on the sites affected by the plant. The nuclear power plant operation caused an increase of tritium activity concentrations downstream from the plant. Radionuclides amounts released after the power plant's extension are estimated.

Keywords

Radioactivity; Tritium; Strontium; Caesium; Background; Surface Water; Nuclear Power Plant

Introduction

The main sources of artificial radionuclides in environment are the tests of nuclear weapons in the atmosphere and the Chernobyl accident. Since then, activity concentrations of tritium (^{3}H), caesium 137 (^{137}Cs) and strontium 90 (^{90}Sr) have permanently decreased(Hanslík et al., 2009; Hanslík and Ivanovová, 2010; Maringer et al., 2004, 2009; Smith and Beresford, 2005; Vakulovsky et al., 1994; Zibold et al., 2002).

In 2001, the nuclear power plant Temelín was put into operation in south of the Czech Republic (see FIG. 1). Nowadays, the nuclear power plant operates two pressurized water reactors with output of 1000 MW each. Temelín's waste water is released into the course of Vltava River after the radionuclides purification. At present, preparation of the nuclear power plant extension is in process. It is expected that the power output of the plant should be doubled: two new blocks, similar to the existing ones, are planned to be constructed in future. As a result, the waste water releases are expected to be about twice larger compared to current state. Launching of the pilot operation of the new blocks is scheduled to 2020 (Web presentation of power plant operator CEZ Group). The aim of this paper is to quantify balances of radionuclides in the area affected by the operation of Temelín nuclear plant and to set the background conditions before the Temelín power plant extension.

Methods

For this study, the data collected by T. G. Masaryk Water Research Institute (TGMWRI) monitoring (Hanslík et al., 1995, 1997, 2000-2012) are used.

The^{90}Srand^{137}Cs activity concentrations were monitored in surface water at several sites at Orlík Reservoir, located at the Vltava River, the Temelín's waste water recipient. These profiles, affected by the current power plant operation, were: Vltava Kořensko and Vltava Solenice. Also sites, not affected by the power plant waste water, were included into the monitoring to set the background conditions. They were located upstream from the waste water outflow (Vltava Hněvkovice) or at tributaries of the Vltava River (LužniceKoloděje and OtavaPísek). The tritium activity concentrations were monitored in LužniceKoloděje, Vltava Hluboká (uninfluenced sites), and at several sites downstream form the waste water outflow. The situation of the

sampling sites is shown on the map in FIG. 1.

In the mentioned sampling sites, the observations of changes in activity concentrations of ^{137}Cs, ^{90}Sr and ^{3}Hwere assessed on surface water.

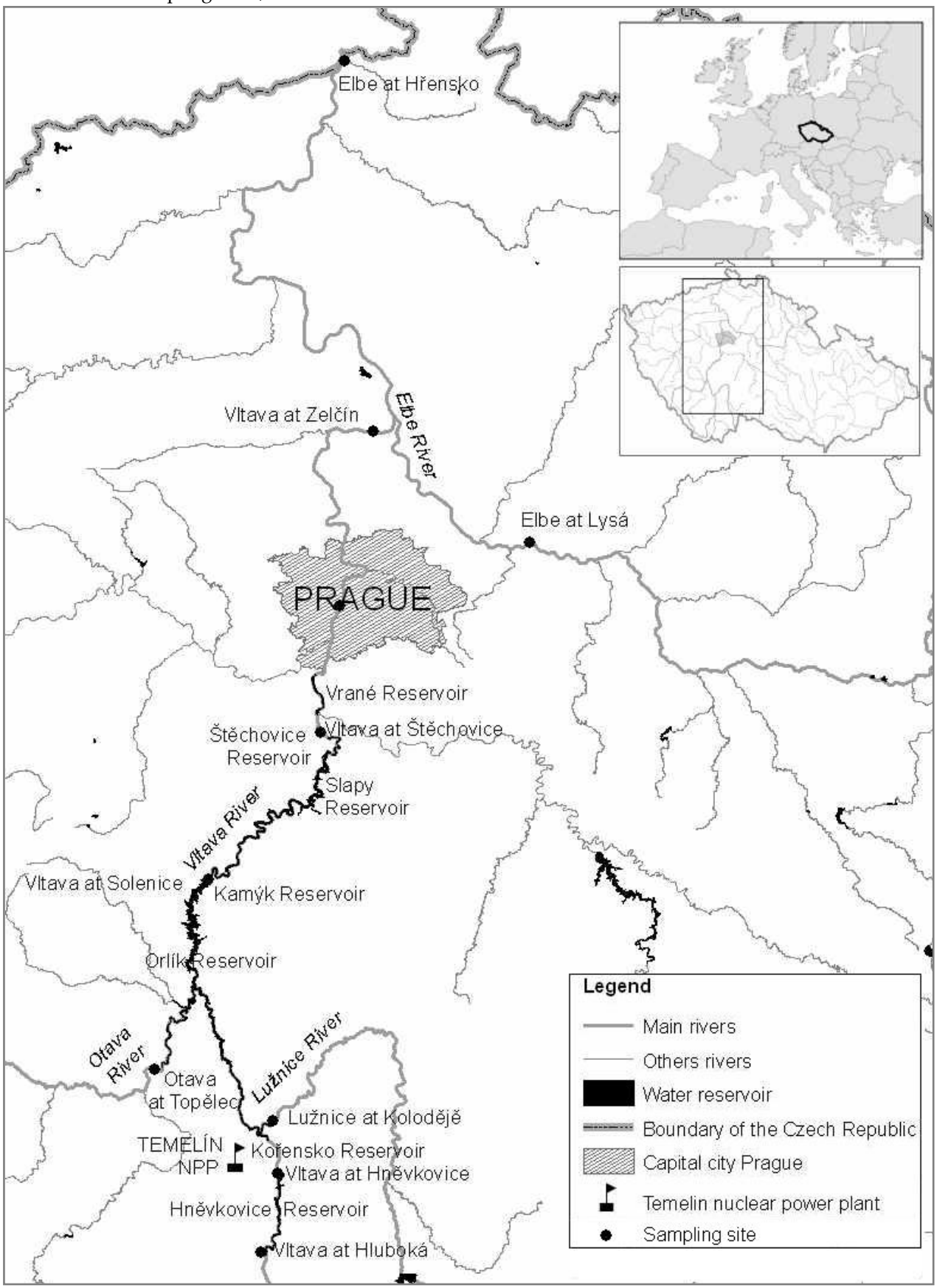

FIG. 1 MAP OF SAMPLING SITES

For sample collection and processing, standardised methods were applied (ČSN EN 25667-1, ČSN EN ISO 5667-3, -4, -6). Quality control practice of the TGMWRI Radioecological Laboratory is performed according to the Standard ČSN EN ISO/IEC 17025. The laboratory takes part in national and international proficiency testing. Calibration is performed by using standards provided by Czech Metrology Institute.

For analysis of ^{137}Cs and ^{90}Sr, large samples of water (50 l) were taken with frequency of four in per year. These samples were immediately stabilized with nitric acid to pH 2 and then, after transportation to the laboratory, dried by vaporization. The vaporized samples were ignited (350°C) and then analysed. Activity concentrations of ^{137}Cs and ^{90}Sr were determined in total solids (both in dissolved and suspended solids).

The ^{137}Cs activity concentrations were analysed according to the standard ČSN ISO 10 703 using gamma-spectrometry. The measurement duration was set up in accordance with the requested minimum detectable activity (MDA) of ^{137}Cs at the level of significance of α=β=0.05. The MDA of ^{137}Cs in water for counting time of 2 days was 0.5 mBq·l^{-1}. ^{90}Sr was determined in water by using a standard method after radiochemical separation (ISO/DIS 13160:2012). To achieve the ^{90}Sr MDA 3 mBq·l^{-1}, the ignited residuum after 50 l water evaporation has to be analysed. Both these methods were verified and recommended by the International Atomic Energy Agency (1996) in Vienna within the framework of its technical assistance organized in co-operation with the Ministry of the Environment of the Czech Republic and the State Office for Nuclear Safety. Samples for ^{3}H analysis (0.25 l) were taken with a frequency of 12 in a year. The ^{3}H activity concentrations were determined by using low-level liquid scintillation spectrometers. The determination was performed according to ČSN ISO 9698. The relative efficiency of tritium measurement was about 25%. The MDA was set according to the expected activity concentrations. For mixture of 8 ml of sample and 12 ml of scintillation solution (Ultima Gold LLT) and alternatively counting time of 800 minutes (for samples not effected by the waste water discharges) and 300 minutes (for samples effected by the waste water discharges), the MDA was 1.2 Bq·l^{-1} and 2.2 Bq·l^{-1} respectively at the level of significance of 0.05.

Our data were completed with data on river flows provided by Czech Hydrometeorological Institute and with data of annual released activities of tritium, ^{90}Sr and ^{137}Cs provided by the CEZ Group that operates the Temelín plant (Fechtnerová, 2001-2006; Lysáček 2007-2012).

The analytical data on the activity concentrations of radionuclides were assessed by using several mathematical methods which are briefly described below.

The effective ecological half-lives were evaluated from the decrease in a radionuclide activity according to the equation (Smith and Beresford, 2005):

$$T_{eff} = \frac{\ln 2}{\lambda_{eff}} \quad (1)$$

where T_{eff} is effective ecological half-life (y), λ_{ef} is effective ecological decay constant of the radionuclide activity concentration (y^{-1}).

Ecological half-lives were calculated by using an equation in the form (Smith and Beresford, 2005):

$$\frac{1}{T_{ecol}} = \frac{1}{T_{eff}} - \frac{1}{T_P} \quad (2)$$

where T_{ecol} is ecological half-life (y), T_P is physical half-life (y).

For a trend analyses, a kinetic equation of the first order was used in the form (an example for ^{137}Cs):

$$\ln c_{137Cs,\,j} = -\lambda_{eff} \cdot t + q \quad (3)$$

where $c_{137Cs,j}$ is annual average ^{137}Cs activity concentration in surface water in year j, in profile k (Bq·m^{-3}); λ_{eff} is effective rate of decline in ^{137}Cs activity concentration (y^{-1}), involving the physical decay constant (λ_P) and the ecological rate of the decrease (λ_{ecol}), $\lambda_{eff} = \lambda_P + \lambda_{ecol}$ (y^{-1}); t is time of the monitoring (y); q is natural logarithm of activity at the beginning of the observation.

Annual balances of assessed radionuclides were calculated according to the equation:

$$A_{ijk} = c_{ijk} . Q_{jk} . t \quad (4)$$

where A_{ijk} is annual balances of radionuclide i in year j in profile k (Bq·y^{-1}); c_{ijk} is annual average activity concentration of radionuclide *i* in surface water in year *j* in profile *k* (Bq·m^{-3}); Q_{jk} means annual river flow in profile *k* in year *j* according to CHMI (m^3·s^{-1}); t is duration of a year (31536000 s)

Values below the MDA were included into the assessment and substituted by 0.5 values of these limits.

Results and Discussion

Background Conditions – Radionuclides in Surface Water

In the beginning of our observation in 1990, the annual

average activity concentration of ^{137}Cs was 10.2 mBq·l^{-1}. The ^{137}Cs activity concentration decreased through years, so the recent results hardly exceeded the detection limits. Nowadays, the detected ^{137}Csactivity concentrations are 1.2 mBq·l^{-1} in average. The ^{90}Sr activity concentrations have decreased from 9.9 mBq·l^{-1} in 1993 to 2.8 mBq·l^{-1} 2012 in average.

The results of temporal changes of ^{137}Cs activity concentrations in the Vltava River and its tributaries were evaluated using kinetic equation of the first order (3). The effective ecological half-lives and ecological half-lives were calculated to describe the radionuclides activity concentrations behaviour in the environment. The effective ecological half-life characterizes the total bulk of various processes and includes also the physical radioactive decay. The ecological half-life represents the environmental processes only. These parameters which are specified in TABLE 1, showed more rapid decrease of the radionuclides activity concentrations in the first years of the observation, as compared with the recent period. Hence, the changes were studied in two periods, 1990-1994 and 1995-2012. The results showed that the evaluated effective ecological half-lives were in range of 1.1–2.2 y in the first period. Against that, they were significantly longer in the second period: the effective ecological half-lives were in range of 6.6–12.9 y. Ecological half-lives were in range of 1.2–2.4 y in the period I and in range of 8.4 –22.6 y in period II.

TABLE 1 PARAMETERS OF KINETICS OF ^{137}CS ACTIVITY CONCENTRATION DECREASE IN SURFACE WATER, ASSESSED IN TWO SEPARATED PERIODS

Sampling site	Vltava Hněvkovice	Lužnice Koloděje	Otava Písek	Inflow to the Orlík Reservoir	Vltava Solenice (outflow from the Orlík Reservoir)
Period I	1990-1994	1990-1994	1990-1994	1990-1994	1990-1994
T_{eff}[year]	1.5±0.9	2.2±1.6	1.1±1.1	1.3±1.3	1.5±0.6
T_{ecol}[year]	1.6±1.0	2.4±1.8	1.2±1.3	1.4±1.4	1.5±0.7
Period II	1995-2012	1995-2012	1995-2012	1995-2012	1995-2012
T_{eff}[year]	6.6±2.3	12.9±7.3	7.9±11.3	8.5±4.6	7.0±2.8
T_{ecol}[year]	8.4±4.1	22.6±22.5	10.6±10.1	11.8±10.5	9.0±5.5

TABLE 2 PARAMETERS OF KINETICS OF ^{90}SR ACTIVITY CONCENTRATION DECREASE IN SURFACE WATER

Sampling site	Vltava Hněvkovice	Lužnice Koloděje	Otava Písek	Inflow to the Orlík Reservoir	Vltava Solenice (outflow from the Orlík Reservoir)
Period	1993-2012	1993-2012	1993-2012	1993-2012	1993-2012
T_{eff}[year]	9.6±3.4	8.2±2.4	10.2±3.4	11.1±3.5	9.4±3.0
T_{ecol}[year]	14.5±7.6	11.4±4.6	15.9±8.2	18.1±9.2	13.9±6.6

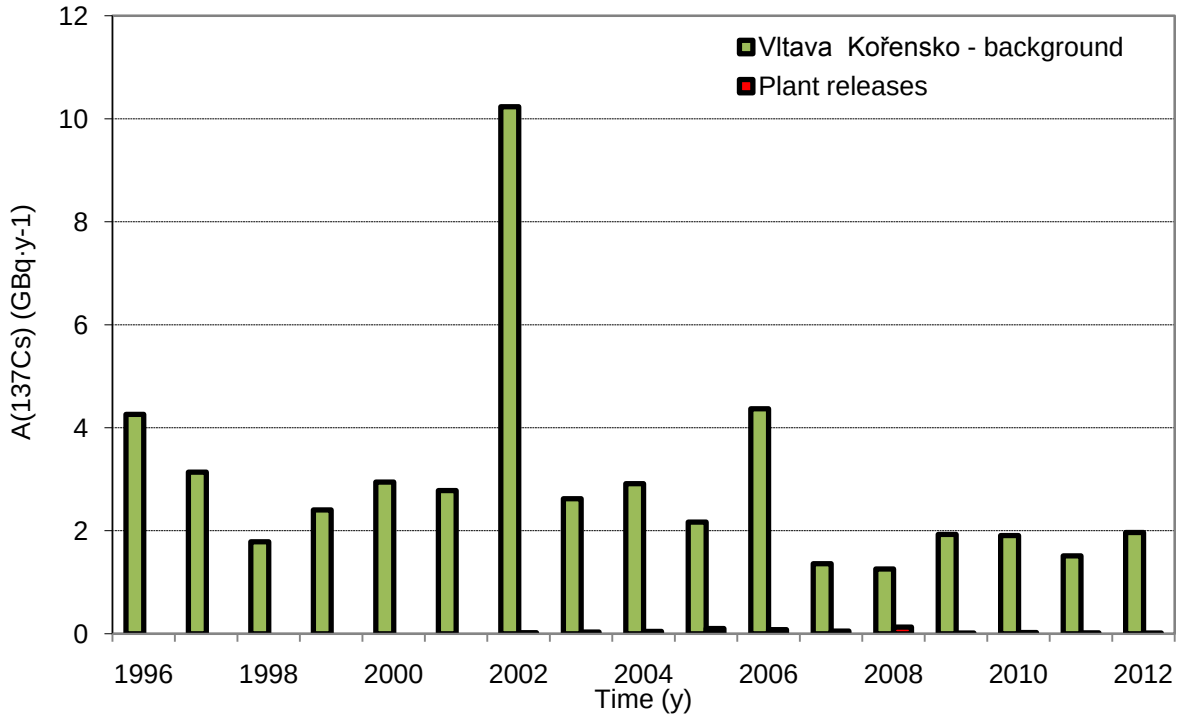

FIG. 2 TEMPORAL CHANGES OF BACKGROUND ^{137}CS BALANCE IN VLTAVA KOŘENSKO AND OF ^{137}CS AMOUNT ANNUALLY RELEASED BY THE POWER PLANT.

^{90}Sr activity concentrations in the Orlík Reservoir and its tributaries were also assessed using a kinetic equation of the first order (3). The results of temporal changes of ^{90}Sr-evaluated effective ecological half-lives and ecological half-lives are in TABLE 3. For ^{90}Sr, the evaluated effective ecological half-lives are in range of 7.5–10.9 y and ecological half-lives are in range of 10.2–17.4 y. In contrast to ^{137}Cs, significant change decrease rate was not observed during the assessed period, so the changes were studied for the whole period of 1993-2012 together for ^{90}Sr.

The differences between effective half-lives determined in particular monitored sites, are due to different conditions which exist in the various environments. They can differ from each other, for example in the flow velocity, suspended solid concentration or the quality of sediments.

For tritium activity concentrations, a slow decrease was observed on surface water in reference sampling site (Vltava Hluboká). This tritium background has three sources:

- a constant component stemming from cosmic radiation,
- tritium, originating from atmospheric nuclear weapons tests, which permanently decreases,
- atmospheric transfer from nuclear facilities worldwide, which is considered to be constant in assessed period.

The decreasing trend in activity concentration of tritium, released in sixties by the nuclear weapons tests, interferes with tritium generated naturally by cosmic radiation and by tritium produced by nuclear facilities worldwide in the monitored period.

Effect of Current Nuclear Power Plant Operation on Hydrosphere

Annual balances of monitored radionuclides were calculated to assess the effect of the currently operating two Temelín's nuclear blocks on the course of Vltava River, recipient of the plant's waste water.

The balances of ^{137}Cs were calculated using the annual average activity concentrations and annual average flow rates in individual sampling sites according to the equation 4. To estimate the radioactive background in the Kořensko profile which is closed to the waste water outflow, the sum of ^{137}Cs balances in Vltava Hněvkovice and Lužnice Koloděje was calculated, then compared with the ^{137}Cs balance released from the Temelín power plant according to the data from the plant operator, CEZ Group, which is shown in FIG. 2. It is obvious that ^{137}Cs residual contamination from atmospheric nuclear weapons tests and Chernobyl accident in the last century is dominant to the activity of ^{137}Cs, released by the nuclear power plant.

The balances of ^{137}Cs are more significantly affected by hydrological situation and behaviour of the background^{137}Cs than by the amounts of^{137}Cs released from the power plant. The maximal ^{137}Cs balance in Vltava at Kořensko was observed in 2002 when extreme floods occurred. On the contrary, in 2003 when serious draughts occurred smaller balance was assessed. The fate and transport of ^{137}Cs is related to activity concentrations of suspended solids in surface water, to which ^{137}Cs is strongly bound. The Orlík Reservoir, located at the Vltava River downstream from the Temelín plant, has a remarkable importance for ^{137}Cs transport, considering the suspended solids sedimentation.

^{90}Sr which is also monitored in the vicinity of the Temelín plant, has different behaviour compared to^{137}Cs. The ^{90}Sr activity concentration is not affected with the Orlík Reservoir by far, since it is not so closely connected to the suspended solids behaviour. Insignificant increase of ^{90}Sr was observed in the outflow from the Orlík Reservoir (sampling site Vltava Solenice), which is attributed to desorption from suspended solids and bottom sediments. ^{90}Sr has significantly lower distribution coefficient (K_D) than ^{137}Cs. According to IAEA (2010) the field distribution coefficients for freshwater ecosystems for ^{90}Sr are in range $2.3x10^2$–$6.3x10^3$ $l \cdot kg^{-1}$ and for ^{137}Cs in range $1.6x10^3$ – $5.2x10^5$ $l \cdot kg^{-1}$.

The amount of ^{90}Sr annually released by the plant is negligible compared to the background^{90}Sr balances in Vltava Kořensko, calculated as the sum of ^{90}Sr balances in Vltava Hněvkovice and Lužnice Koloděje (data from monitoring). It is obvious that ^{90}Sr residual contamination from atmospheric nuclear weapons tests and Chernobyl accident in the last century is dominant as compared to the ^{90}Sr amount released by the plant.

As illustrated in FIG. 3, the tritium activity concentrations have increased in Vltava River downstream from the Temelín waste water outflow since the Power plant has been put into operation in 2001.The maximal annual average tritium activity concentration was observed at sampling site Vltava Solenice (26.1 $Bq.l^{-1}$) in 2011.

The tritium annual average activity concentrations in Vltava Solenice and in reference site Vltava Hluboká

and annual average river flows were used for calculation of tritium balances, then compared to the ^{3}H released from the power plant. The results are in FIG. 4. The total released amount of ^{3}H, calculated as a sum of individual annual balances for the period 2001-2012, is 427TBq according to the surface water observation, which is in a good agreement with 405TBq stated by CEZ Group.

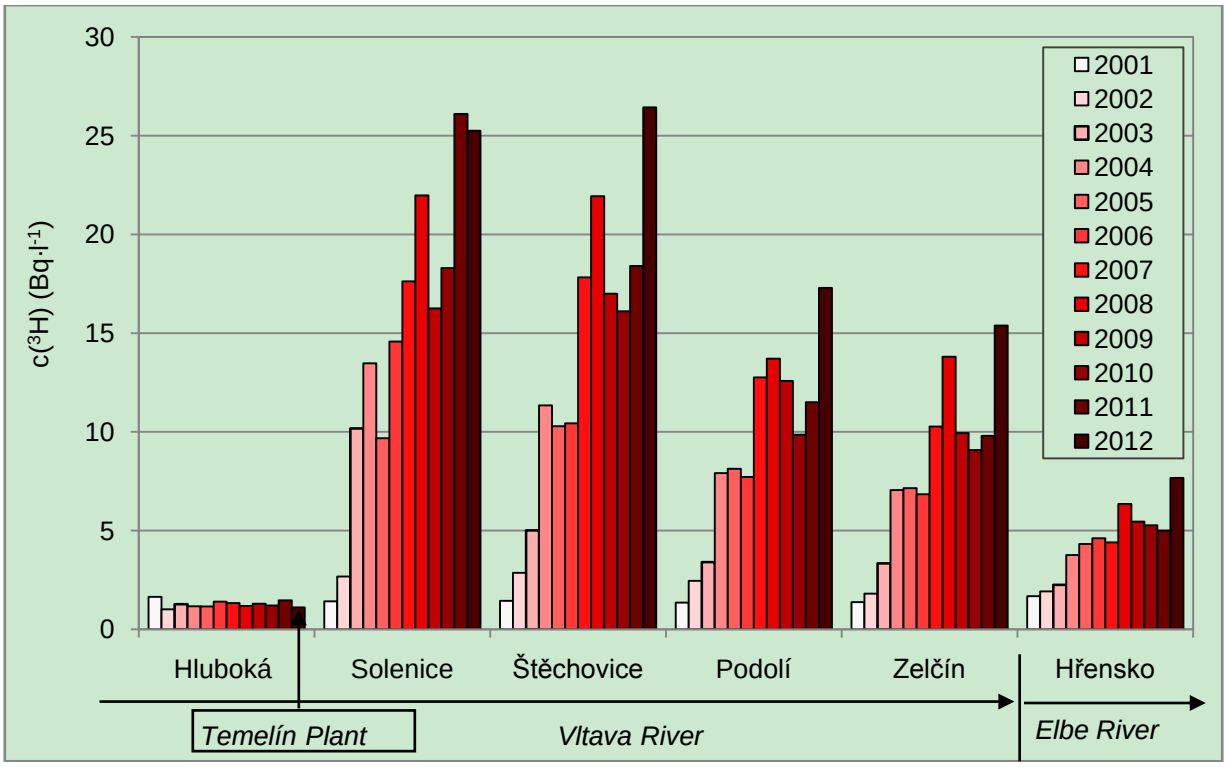

FIG. 3 ANNUAL AVERAGE TRITIUM ACTIVITY CONCENTRATIONS IN THE VLTAVA RIVER UPSTREAM (HLUBOKÁ) AND DOWNSTREAM FROM THE OUTFLOW OF TEMELÍN PLANT IN 2001 – 2012.

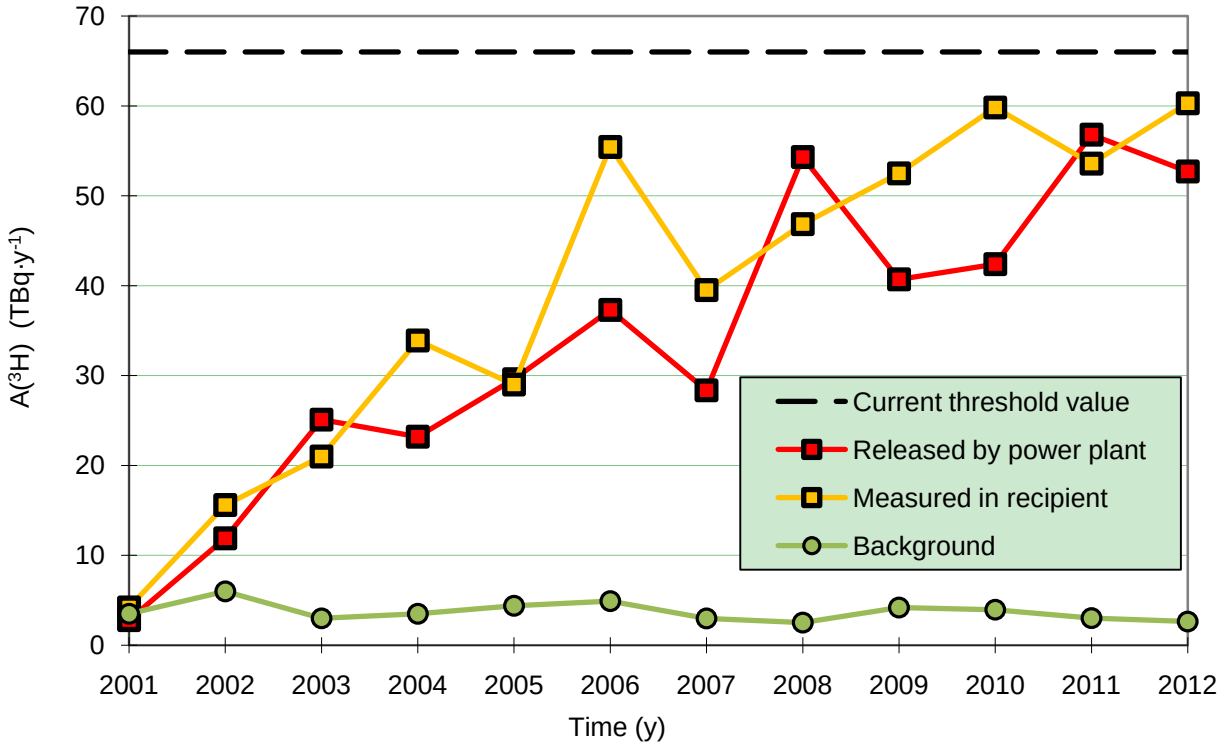

FIG. 4 COMPARISON OF ANNUAL TRITIUM BALANCES IN THE TEMELÍN'S RECIPIENT (VLTAVA SOLENICE), BACKGROUND ^{3}H BALANCE (VLTAVA HLUBOKÁ) AND ANNUALLY RELEASED TRITIUM ACCORDING TO THE DATA PROVIDED BY THE CEZ GROUP.

Prognosis of the Power Plant Extension Influence on Hydrosphere

For the estimation of the future impact of the power plant extension, the experience from the recent power plant operation was utilized. After the first recently operated, two blocks start-up in 2001, the released tritium amount steadily grew until 2008. Then, annually released tritium amounts have fluctuated around a stable value. A similar schedule of launching and the pilot operation was supposed also for the launching of the new source, implying that the presumption of the tritium released amounts increases in the first years after the new blocks start-up and the amount of tritium released during stabilized full operation that follows. The estimation of the ^{3}H amounts, released by the Temelín Power plant, is in FIG.6. The current threshold value for annually released tritium is 66 TBq. After the power plant extension, the threshold is expected to raise in consequence of the increase of the power plant output.

If the suggested threshold value for annually released tritium (132 $TBq \cdot y^{-1}$) is fulfilled, the predicted annual average tritium activity concentrationin Kořensko, the profile closed to waste water release, is about 84 $Bq \cdot l^{-1}$.The annual average flow rate is 50 $m^{3} \cdot s^{-1}$ at Kořensko. It is presumable that the real ^{3}H average activity concentration will be lower, as the released ^{3}H amount would be lower than the threshold value.

As ^{137}Cs and ^{90}Sr are concerned, their released amounts are also expected to grow twice, as the power plant output is doubled. During 2008–2012, the annual released amount of ^{137}Cs was approximately 40 $MBq \cdot y^{-1}$, for ^{90}Sr, it was 0.4 $MBq \cdot y^{-1}$. After the power plant extension, the released amounts are expected to reach approximately 80 $MBq \cdot y^{-1}$ for ^{137}Cs and 0.8 $MBq \cdot y^{-1}$ for ^{90}Sr. These amounts represent only a minor part of the annual background balance in Vltava Kořensko for ^{137}Cs and even less in the case of ^{90}Sr. Even in future, when the ^{137}Cs and ^{90}Sr background activities are expected to decrease, the amounts of ^{137}Cs and ^{90}Sr, released by the extended power plant, are still disguised by the radioactive background, stemming from the nuclear weapons tests and the Chernobyl accident.

Conclusion

On surface water, a decrease of ^{137}Cs and ^{90}Sr activity concentrations has been observed upstream and downstream from the outflow of waste water from Temelín plant. The influence of the Temelín plant on the ^{137}Cs and ^{90}Sr activity concentrations was almost completely covered by the residual contamination from global fallout and the Chernobyl accident. In case of tritium, very slow decrease of background activity concentrations was observed on reference sampling sites. Downstream from the plant, significantly higher activity concentrations were detected corresponding to tritium discharged from Temelín.

After the future power plant extension, the released amounts of radionuclides are expected to be doubled. This increase is significant in the case of tritium, nevertheless the ^{3}H activity concentrations are still expected to be lower compared to the indication parameters for tritium in drinking water set by European Commission (100 $Bq \cdot l^{-1}$) (98/83/EC). On contrary, the released amounts of ^{137}Cs and ^{90}Sr are still negligible, compared to the residual contamination of surface water caused by nuclear weapons tests and the Chernobyl accident.

In connection to the planned power plant extension, it is necessary to continue the monitoring of the current state of the hydrosphere, so the background values are well known.

ACKNOWLEDGMENT

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